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Column for carrying out an isotope exchange between a liquid substance and a gaseous substance

The invention relates to a column for carrying out an isotope exchange between a liquid substance and a gaseous substance using a catalytic reaction. It also relates to uses of the column.

A method is known from US-A- 4 143 123 by means of which an exchange of hydrogen isotopes can be carried out, namely between liquid water  $H_2O$  and hydrogen gas  $H_2$ , in which the hydrogen H ("protium") is partly substituted by deuterium D or tritium T. In this method a central role is played by a reversible reaction in which an isotope which is bound in a hydrogen molecule is catalytically substituted by a hydrogen isotope of a water molecule. This reaction is sufficiently efficient only when the catalytically active material is dry, i.e. remains unwetted by liquid water. A catalytically active material has therefore been developed which is coated with a semi-permeable film which is hydrophobic and hence water repellent on the one hand but which is however permeable for free water molecules from an environment which is formed by water vapor or a moist gas on the other hand.

The object of the invention is to create a column in which for example an exchange of hydrogen isotopes between liquid water and hydrogen gas can be carried out and in which measures are realized, as a result of which a contamination or wetting of the catalyst by the liquid does not occur. In addition the column should be free of dead spaces. This object is satisfied by the column which is characterized in claim 1.

The column is provided for carrying out an isotope exchange between a liquid substance and a gaseous substance using a catalytic reaction. In this reaction the isotope exchange between molecules of the vaporized liquid substance and molecules of the gaseous substance takes place through a heterogeneous catalysis. The column comprises a plurality of modules which are arranged vertically one above the other and which are in each case subdivided into two regions K and A which are serially connected by a connection region. The catalytic reaction can be carried out in the region K on a first packing. A substance exchange between a liquid and a gaseous phase which contains vapor can be carried out in the region A by means of a second packing for compensating substance concentrations. During the operation of the column a transport of the gaseous substance which contains vapor is driven through the modules as a result of pressure gradients. In this the transport direction is changed at least once in the connection region, and indeed from a downward direction to an upward direction, whereas the liquid substance is forwarded downwardly through the modules through the action of gravity alone.

Subordinate claims 2 to 8 relate to advantageous embodiments of the column in accordance with the invention. Two possible uses of the column in accordance with the invention are in each case the subject of claim 9 and claim 10.

In the following the invention will be explained with reference to the drawings. Shown are:

- Fig. 1            a tabulation of abbreviating designations as well as relationships relating to the isotope exchange,
- Fig. 2            part of a longitudinal section through a column in accordance with the invention,
- Fig. 3            a diagram in which changes of substance concentrations for a module of the column are qualitatively illustrated,
- Fig. 4            a schematic illustration of a plant with a nuclear reactor and a device for the de-enrichment of tritium from heavy water,
- Fig. 5            relationships pertaining to a further example of an isotope exchange,
- Fig. 6            a module pertaining to a column in accordance with the invention which has a centrally symmetrical construction and

Figs. 7, 8 two further variants of the module.

The isotope exchange, for the carrying out of which the column in accordance with the invention is provided, takes place between a liquid L and a gas G, with it being necessary for the liquid L to be present as a vapor V for a catalytic partial step of the isotope exchange. The liquid L is for example heavy water  $D_2O$  in which a certain proportion of its molecular components has a different composition in regard to the isotopes: in  $D_2O$  ( $=L_{22}$  in accordance with the relationships tabulated in Fig. 1, columns 1 and 1a) a deuterium D is substituted by tritium T, so that DTO ( $=L_{23}$ ) forms a second component. Other examples are also possible in which L consists of a different mixture of two or more of the components  $L_{ij}$  (with the indices  $i, j = 1, 2$  or  $3$ ), which are tabulated in Fig. 1. The vapor D of the liquid L has a corresponding composition of components  $D_{ij}$  ( $i, j = 1, 2, 3$ ), the meaning of which can be found in the columns 1 and 1b in Fig. 1. The gas G consists analogously of components  $G_{ij}$  ( $i, j = 1, 2, 3$ ), the meaning of which is given in the columns 2 and 2b. It should be observed that the composition of the three isotopes H, D and T can be read off on the basis of the indices  $i$  and  $j$ : the index 1 corresponds to a H, the index 2 to a D and the index 3 to a T.

In Fig. 1 relationships by means of which the isotope exchange is symbolically illustrated are additionally given for an example. The relationship EM relates to the reversible isotope exchange between  $L_{23}$  and  $G_{22}$  in the one reaction direction or between  $L_{22}$  and  $G_{23}$  in the

reverse reaction direction; the relationship EM thus describes the exchange of T and D between heavy water and heavy hydrogen. This exchange EM, which relates to a module M of a column 1 in accordance with the invention (cf. Fig. 2), takes place via physical-chemical partial steps, namely exchange processes which take place in two separate regions A and K of the module M and which can be represented by the relationships E1, EK and E2. E1 and E2 are reversible aggregate state changes of water molecules, namely the vaporization of the components  $L_{23}$  and  $L_{22}$  respectively and the condensation of the components  $V_{22}$  and  $V_{23}$  respectively. In these exchange processes there results a substance exchange between a liquid and a gaseous phase containing vapor, which leads to a compensation of oxygen concentrations. The relationship EK represents the catalytic reaction which takes place in the region K of the module M: molecules of the gas G and of the vapor V are adsorbed on a catalytically active material and are again desorbed from the latter, with the isotope exchange taking place with a certain probability in the adsorbed state.

The column 1 which is partly illustrated in Fig. 2 comprises a plurality of modules M which are arranged vertically one above the other. These are in each case subdivided into two regions K and A which are serially connected by a passage 4 or connection region C. In the region K the catalytic reaction EK takes place on a packing 2 which is flowed through by the gas G containing the vapor V. In the region A a substance exchange which is based on the state changes E1 and E2 between the liquid L and the gas flow consisting of G and V is produced by means of a packing 3. After passage through the packing 3 the liquid

L is conducted further via a collector 5 to a distributor 51 of the adjacent module. The liquid collector 5 contains a siphon-like drain 50 by means of which a upward passing through of gas can be prevented.

The transport of the gas flow consisting of G and V is driven by pressure gradients. The gas flow passes upwardly in the region K, downwardly in the connection region C, and upwardly again in the region A. The liquid L is forwarded downwards through the modules M of the column 1 by gravity alone.

A filling of porous particles, on the surface of which a catalytically active material is applied, can be used as a packing 2. Examples of catalysts are given in the already named US-A- 4 143 123. For the substance exchange in the region A an ordered packing 3 can be used. This latter is in particular built up of vertical layers which contain inclined channels which are produced from corrugated material surfaces and which thereby form a cross channel structure with channels which cross openly. The packing 2 can also be formed as an ordered packing with a cross channel structure in which a wash coat is used as a carrier of the catalytic material. The packing 3 can correspondingly be formed of a filling of packing bodies. The packing 3 is advantageously manufactured of copper, since a copper oxide film which is easily wettable can be produced on copper.

The architecture of the column 1, which is schematically illustrated in Fig. 2, has the following advantages: There are no detrimental dead spaces, since all spatial regions are flowed through. The two regions A

and K are screened off from one another in such a manner that no liquid drops can pass from A to K. Droplets of this kind would on the one hand impair the efficiency of the catalysis in K; on the other hand copper would be transported by the droplets, which would contaminate the catalyst and thus reduce its reactivity.

Fig. 3 shows with reference to a diagram how the concentrations of the components  $G_{ij}$ ,  $V_{ij}$  and  $L_{ij}$  vary as a result of the exchange processes E1, EK and E2 (see Fig. 1) in a module M of the column 1. The variations are illustrated only qualitatively. The couplings of the concentrations as a result of the exchange processes E1, EK and E2 are made recognizable with broken connection lines. The curve sections are illustrated as arrows in order to make the directions of movement of the components  $G_{ij}$ ,  $V_{ij}$  and  $L_{ij}$  through the regions A, C and K visible.

Fig. 4 shows a schematic illustration of a plant 7 with a nuclear reactor R in which heavy water (= L with the component  $L_{22}$ ) is used as a moderator and cooling water. Through neutron capture D is converted into T in the heavy water. In the column 1 in accordance with the invention the tritium is de-enriched from the heavy water (= L with the components  $L_{22}$  and  $L_{23}$ ) by means of a gas flow (= G with the component  $G_{22}$ ) consisting of D<sub>2</sub>. The gas flow (= G with the components  $G_{22}$ ,  $G_{23}$  and  $G_{33}$ ) which is charged with tritium is regenerated in a low temperature column 6, with tritium being separated off (in the form of  $G_{23}$  and  $G_{33}$ ).

The column in accordance with the invention can also be used for winning heavy water. The exchange processes E1', EK' and E2' are summarized in Fig. 5. In this method a hydrogen gas which contains D forms the gaseous substance G and a mixture of light and heavy water forms the liquid substance L. Deuterium is given off from G to L; at the same time hydrogen (i.e. protium) is taken up from L by G.

Fig. 6 shows a module M pertaining to a column in accordance with the invention which is formed as a boiler shell ring or pipe length 10 and which is formed centrally symmetrically. The packing 3 for the substance exchange E1 and E2 is located in a cylindrical chamber 3' which forms the region A. The connection region C comprises a first ring space 4, which is arranged concentrically to the region A. The region K with the packing 2 is formed by a second concentric ring space 2'. The liquid L is distributed onto the packing 3 by a cylindrical distributor 51.

Fig. 7 shows a further centrally symmetrical module in which the region K is located in a ring space 2" which is arranged directly around the central region A. An outer ring space 4' forms a connection region between adjacent modules M. In the connection region C between the regions A and K, which is no longer formed as a separate partial space in this example, the transport direction of the gas flow which consists of G and V is changed only once: After a flowing through the region K — downwardly — the flow changes on passage through the connection region C into the reverse direction, which is directed upwards. Thanks to this change of direction there is no danger that droplets pass from the region A into the region K.



The further embodiment in Fig. 8 — illustrated as longitudinal section and cross-section — has a region K which is composed of a plurality of partial regions K1, K2, K3 and K4 which are arranged in parallel. The space between these partial regions forms a part of the connection region C. The packing 3 of the region A is arranged in a space having the shape of a rectangular parallelepiped.

FIG. 8